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13. ABSTRACT Maximum 200 words)

Vacuum ultraviolet radiation from rocket plumes offers a unique high-altitude diagnostic observable for identifying and tracking incoming missiles. The interaction of rocket motor combustion products C2H2, C2O, and C2 with the atmospheric constitutents O and O2 was shown to lead to CO(A-X) (4+) chemiluminescence. The spectra and kinetics of the reactions were studied in a heatable pseudo-static photochemical reactor and in fast-flow reactors. The O + C2O and C2 + O2 reactions were shown to proceed through excited C2O2 intermediates. The chemiluminescence from C2 + O2 was used as the diagnostic for rate coefficient determinations; the results compared well to laser-induced fluorescence measurements, which led to k(293-1250 K) = 1.1 x 10(-11)exp(-392 K/T) cm(3) molecule(-1)s(-1). In addition to 4+ radiation, the C2 + O2 reaction produced an apparent continuum in the vuv, probably due to C2O2 radiation to a repulsive ground state. This suggests the possibility for developing a chemical laser for the vuv.

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Research Objectives

The purposes of this work were to obtain kinetic information on chemiluminescent reactions for incorporation in missile plume radiation codes and to train students in gas-phase chemiluminescence laboratory techniques.

Results and Publications

The progress made is best described by the publications resulting from the grant.

1. Arthur Fontijn, Abel Fernandez, Aleksandra Ristanovic, Mai Y. Randall, and Jerome T. Jankowiak, "CO Chemiluminescence and Kinetics of the C₂ + O₂ Reaction" The Journal of Physical Chemistry 105, xxxx (5 April 2001)

The reaction $C_2(X^1\Sigma, a^3\Pi) + O_2(X^3\Sigma) \rightarrow CO(X^1\Sigma, A^1\Pi, a^{3}\Sigma, d^3\Delta, e^3\Sigma) + CO(X^1\Sigma)$ has been studied in two types of reactors. A Pyrex steady-state fast-flow reactor at approximately 500 K was used to obtain the spectral distributions of the CO ($A^1\Pi$ - $X^1\Sigma$) emissions in the vacuum ultraviolet (vuv) and the CO triplet states emissions in the visible and near ir (vis) wavelength region. The vuv emission had not previously been postively identified. The C₂ was produced from the C₂Cl₄ + K reaction. In a pseudo-static high-temperature photochemistry (HTP) reactor C₂ was made by 193 nm multiphoton dissociation of C₂Cl₄. That apparatus was used for quenching and rate coefficient experiments in the time domain. The vuv quenching measurements confirm the orbital symmetry argument that the reaction proceeds through excited C_2O_2 intermediates. Reaction schemes for $C_2(X^1\Sigma)$ and for $C_2(a^3\Pi)$ are presented. The results are compared to those from the $O + C_2H_2$ reaction, which leads via $C_2O + O$ to the same band systems emissions, and the differences are discussed below (Paper 2). Measurements of the overall rate coefficients from the decrease of the vis chemiluminescence intensities with time yielded: $k_{vis}(298-711 \text{ K}) = 1.1 \times 10^{-11} \exp(-381 \text{ K/T}) \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$, with 2σ precision limits of around \pm 5% and corresponding estimated accuracy limits of about \pm 21%. This expression is in excellent agreement with earlier rate coefficient values determined by different methods, which leads to the recommendation $k(293-1250 \text{ K}) = 1.1 \times 10^{-11} \exp(-392 \text{ K/T})$. The vuv experiments yielded slightly higher values, the reason for which is uncertain. It is speculated that an apparent continuum observed in the vuv spectra could be ¹C₂O₂ excimer radiation to the repulsive ground state. This suggests the possibility of using the $C_2 + O_2$ reaction to produce a chemical laser for vacuum ultraviolet wavelengths.

2. Arthur Fontijn, Abdellatif Goumri, and Paul E. Brock II, "Pressure-Dependence of the $CO(d^3\Delta - a^3\Pi)$ Triplet Bands Chemiluminescence Intensities from the $O + C_2H_2$ Reaction: Mechanistic Implications", Combustion and Flame, 121, 699 (2000).

It is shown that the (visible) Triplet Bands of CO from the $O + C_2O$ reaction decrease in intensity with increasing pressure. This behavior is opposite to that of the (vuv) CO 4+ bands, which increase in intensity. This indicates a mechanism where the triplet emitter molecules form first and by collision-induced cross-relaxation produce the singlet emitters. By contrast the CO 4+ bands from $C_2 + O_2$ decrease in intensity with increasing pressure and the cross-relaxation mechanism does not contribute significantly there, as further discussed in Paper 1.

Theses

و المراجع

The following theses have resulted in part from support under the present grant:

M.Y. Randall, "Kinetics of the CO Fourth Positive Chemiluminescence from the $C_2 + O_2$ Reactions", M.Sc., Rensselaer Polytechnic Institute, Troy, NY (1998).

A.S. Blue, "Temperature Dependence of the Kinetics of Transient Species Reactions", Ph.D., Rensselaer Polytechnic Institute, Troy, NY (2000).

Further Students Supported by this Grant

In addition to the two above US students who have completed their theses, one ongoing Ph.D. student, Abel Fernandez, and a dropped-out graduate student Jessica Kasner, participated. A number of Rensselaer US undergraduates also received support under this grant and participated for periods varying from 3 to 12 months. These people are: Jerome T. Jankowiak, Ryan Youngsaye, Todd Alexander, Charles Fung, Christine Brown, Erin Carhart, Inuka Dissanayake, and Michele Nardone. Partially as a result of this first research experience, several of these students decided to go to graduate school.